Third United Nations International Conference on the Peaceful Uses of Atomic Energy

A/CONF.28/A/798
JAPAN
February 1964
Submitted in:
ENGLISH and FRENCH

Confidential until official release during Conference

Abstract

Chlorination-Distillation Processing of Irradiated Uranium Dioxide and Uranium Dicarbide

by Takehiko ISHIHARA and Kenmei HIRANO
Japan Atomic Energy Research Institute

Chlorination-distillation processing of irradiated pellets of uranium dioxide and of uranium dicarbide was studied, with the aim of utilizing the difference in vapor pressure between chlorides of uranium and those of fission products. The condensed uranium chlorides were recovered in the form of dioxide powder by oxidation-reduction cycling.

Pellets were irradiated during either 1 or 4 weeks in the JRR-1 with an intergrated flux of 0.25×10^{16} or 1×10^{16} n/cm^2 and were subjected to experiment after letting the activity decay for a period of between one and five weeks. The dioxide pellets were chemically pulverized by alternately oxidizing in air flow at about 400°C and reducing in hydrogen flow at about 800°C, while the dicarbide pellets were used without pulverization. Carbon tetrachloride was used as chlorinating agent, in the form of vapor flowing through a quartz-tube assembly at various chlorination and condensation temperatures.

It was not possible to obtain the separation of most of the fission products from irradiated uranium dioxide by a simple

process of chlorination-fractional distillation followed by oxidation-reduction, and the decontamination factor (γ) was only about 10 with fuel cooled 40 days. The γ-emitting nuclides remaining with the recovered uranium were mainly ^{141}Ce, ^{144}Ce and ^{239}Np. ^{239}Np decays rapidly due to its short half-life. In order to separate the radioactive cerium, natural cerium dioxide powder was added to an amount corresponding to 2 o/o of the irradiated uranium dioxide before chlorination-distillation, which was followed by the oxidation-reduction cycle. The decontamination factor obtained in this case was approximately 100 with fuel cooled 40 days. By repeating this process with cerium dioxide addition, the decontamination factor was further raised to about 400, with more than 97 o/o of uranium recovered.

The uranium dicarbide pellets were directly chlorinated without pulverizing, and distillated fractionally immediately after chlorination. Carbon remained involatile, retaining most of the cerium. The decontamination factor (γ) obtained was approximately 150 on fuel cooled 40 days. For better decontamination, a second chlorination-fractional distillation was conducted after adding 2 o/o cerium dioxide, followed by the oxidation-reduction cycle, as on the irradiated uranium dioxide. The decontamination factor of the recovered uranium dioxide was about 900. More than 95 o/o of uranium was recovered.